# CHAPTER VI
# CONCLUSION AND RECOMMENDATIONS

Barium strontium titanate filled polybenzoxazine based composites were proposed as new dielectric materials operated at microwave frequency. To alleviate the brittle nature of usual monomeric benzoxazines, in this work, the toughness of polybenzxoazine was improved via two strategies. The first method is by alloying with flexible urethane resin. Another strategy is by utilizing a synthesis route of high molecular weight benzoxazine precursors. The effect of different molecular structures was studied based on dielectric characteristics including thermal and mechanical properties. The appropriate polymer matrix provided several outstanding properties of each preparation route was used for composite fabrication

For the first approach, the combination between aniline-based poly benzoxazine and urethane elastomer yielded several superior properties such as thermal, mechanical and electrical characteristics. The incorporation of small amount of polyurethane could help increase decomposition temperature, flexibility, glass transition temperature and also dielectric properties. At 90/10 vol.% of poly(benzoxazine/urethane), (BA-a/PU) was outstanding in dielectric properties. Thus it was used as a polymer matrix. By using barium strontium stitanate, $Ba_{0.3}Sr_{0.7}TiO_3$, (BST) as ceramic filler, the dielectric of the composite was improved owing to high dielectric constant in its nature with low loss tangent. The adding BST filler was not only help enhance dielectric constant but also decrease the dissipation factor. Using 3-aminopropyl trimethoxy silane and BA-a/PU polymer base as surface modifiers on BST could help improve the dielectric constant and filler distribution. At 60 wt.% of silane treated BST exhibited the highest dielectric constant (13.9) with low loss tangent (0.0095) observed at room temperature (1 GHz).

As another method, main chain benzoxazine polymer (MCBP) approach by the reaction of bisphenol A, paraformaldehyde, and diamine provided more ductile cross-linked polybenzoxazines and higher toughness. These lead to prevent the termination of chain propagation and intramolecular hydrogen bond formation, which are the drawbacks of using small molecular weights benzoxazine monomers.

The dielectric characteristics of high molecular weight polybenzoxazines were investigated for the first time at microwave frequencies (300 MHz to 1 GHz) from -50 °C to 150 °C. It was found that the dielectric behaviors strongly depended on the molecular structure of the polymer. A higher dielectric constant and loss tangent were found in methylenedianiline-based (BA-mda) polybenzoxazine containing an aromatic unit from diamine-based. The minor dielectric variation of poly(BA-mda) appeared above 50 °C. With aliphatic in the main chain, hexamethylenediamine-based (BA-hda) polybenzoxazine, showed a lower relative permittivity and dissipation factor owing to the lower polarizability of saturated hydrocarbon bonds. Therefore, poly(BA-mda) was used as a polymer matrix. For poly(BA-mda) composites, the dielectric constant was slightly increased in relation to the amount of BST loading. Surface modification route by silane coupling agent and BA-mda rarely improved dielectric characteristics and the dispersion of ceramic filler. At 60 wt.% of silane treated BST provided the dielectric constant and loss tangent around 8.2 and 0.0268, respectively.

According to the theoretical model prediction, BA-a/PU composites were fit well with Lichnecker model at low amount of BST indicated that the composites were a random mixture of nearly spherical inclusion. For higher BST content, Maxwell-Wagner equation was practical for predicting the relative permittivity of the composites suggesting that uniformly distributed spherical inclusions of ceramic material with dielectric constant. While, poly(BA-mda) composites were likely to follow Maxwell-Wagner equation.

Using poly(benzoxazine/urethane) alloyed and main chain benzoxazine polymer could improve the brittleness confirmed by the exhibiting larger plateau region than that of aniline-based polybenzoxazine. The dielectric properties of all compositions were found to be closely a linear relationship in the wide temperature range and frequency observed when compared with other polymers implying low relaxation behaviors. Besides, the polymer composites had high thermal stability.

It is proved that the proposed polybenzoxazine based composites could be new dielectric materials developed for microwave frequency applications.

**Recommendations**

1. For composite preparation in high molecular weight polybenzoxazine based, the new type of conductive fillers should be introduced instead of ceramic filler so as to shift dielectric constant to higher value and to eliminate the large different in density causing the phase separation between the two components.

2. The other types of surface modifications containing the aromatic bulky group or epoxy silane which is similar to benzoxazine structure should be applied to enhance the dispersion of ceramic filler by taking the advantage of identical structure.

3. With respect to the appearance of undesirable phase (Ba or $SrCO_3$), the calcination temperature should be concerned. Because this parameter also affects to the particle size, which has a direct impact on dielectric properties, the study on temperature would be optimized thoroughly.

4. To increase the quality factor of the composites, the loss factor is important. As a result, barium strontium titanate should be doped with other metals such as MgO and MnO in order to decrease the dielectric loss.

5. The study on dielectric properties at low frequency should be carefully performed because the technical error of the instrument was found during measurement resulting in obtaining a very high dielectric constant over the actual value of the prepared materials. Thus the compensation program should be applied for this work.